



Atmospheric aerosol deposition influences marine microbial communities in oligotrophic surface waters of the western Pacific Ocean



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ABSTRACT

Atmospheric aerosols contain particulates that are deposited to oceanic surface waters. These can represent a major source of nutrients, trace metals, and organic compounds for the marine environment. The Japan Sea and the western Pacific Ocean are particularly affected by aerosols due to the transport of desert dust and industrially derived particulate matter with aerodynamic diameter less than 2.5 μm (PM_{2.5}) from continental Asia. We hypothesized that supplementing seawater with aerosol particulates would lead to measurable changes in surface water nutrient composition as well as shifts in the marine microbial community. Shipboard experiments in the Pacific Ocean involved the recovery of oligotrophic oceanic surface water and subsequent supplementation with aerosol particulates obtained from the nearby coastal mountains, to simulate marine particulate input in this region. Initial increases in nitrates due to the addition of aerosol particulates were followed by a decrease correlated with the increase in phytoplankton biomass, which was composed largely of Bacillariophyta (diatoms), including *Pseudo-nitzschia* and *Chaetoceros* species. This shift was accompanied by changes in the bacterial community, with apparent increases in the relative abundance of heterotrophic Rhodobacteraceae and Colwelliaceae in aerosol particulate treated seawater. Our findings provide empirical evidence revealing the impact of aerosol particulates on oceanic surface water microbiology by alleviating nitrogen limitation in the organisms.

1. Introduction

Aerosols transported by westerly winds are a major source of nutrients, trace metals, and organic matter in the western Pacific Ocean (Jo et al., 2007) and Japan Sea (Tan et al., 2013). In oceanic areas where photosynthetic and heterotrophic microbial communities are nutrient-limited, aerosol deposition can supply both necessary trace elements and nutrients. The subsequent growth of primary producers affects marine productivity and carbon sequestration (Blank et al., 1985; Prospero and Savoie, 1989; Duce and Tindale, 1991; Spokes and Jickells, 1996; Erickson et al., 2003; Yuan and Zhang, 2006). Changes in the dynamics of bacterial communities induced by aerosol deposition to oceanic areas are also thought to vary the mineralization of organic matter and therefore affect marine

carbon cycles (Lekunberri et al., 2010; Romero et al., 2011).

Recent economic and industrial development in continental Asia, particularly in China, has increased the emission of air pollution from the mainland coast (Chan and Yao, 2008). The deposition of anthropogenic aerosols from industrial regions delivers nutrients, such as nitrate and phosphate, to the ocean, significantly enhancing marine biological productivity along the Pacific Northwest coast (Zhang and Liu, 1994; Tan et al., 2011). Interestingly, trace elements, such as copper (Cu), found in dust particles have been reported to inhibit microbial growth in aquatic environments (Paytan et al., 2009; Jordi et al., 2012). Aerosol deposition inputs nutrients and trace elements to oligotrophic oceanic surface waters. The atmospheric input of nutrients into the North Pacific Ocean increases during short-term episodic deposition events during the high dust season (DiTullio and Laws,

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1991). In the North Pacific Ocean, one large dust event is found to have deposited aerosol particulates onto surface waters at a rate of approximately 6.0 mg L^{-1} one-event⁻¹ (Duce and Tindale, 1991). The annual deposition of dust generally varies between the Japanese coastal area (21 g m^{-2}) and the open ocean (0.8 g m^{-2}) (Uematsu et al., 1985). Deposited aerosols remain suspended within 40 m of the surface for up to 72 h (Hashimoto et al., 2005).

Dust events are also known to transport airborne microorganisms, supporting microbial recruitment to ecosystems downwind (Hervás et al., 2009). Although previous studies have analyzed the influence of aerosols on marine ecosystems using model simulations and chemical analyses, there have been few incubation experiments that have directly assessed the response of marine microbial communities to aerosol deposition (Lekunberri et al., 2010; Romero et al., 2011). Incubation experiments using aerosol-treated seawater samples are affected by several constraints. Some studies have investigated microbial dynamics in marine and lake water samples treated with dust particulates and incubated under laboratory conditions (Paytan et al., 2009; Lekunberri et al., 2010). However, the time required for the transfer of open ocean seawater samples to the laboratory affects the microbial community. Shipboard incubation experiments can overcome this issue. Another challenge consists of collecting the large volumes of aerosols required to perform seawater incubation experiments.

During the winter and early spring, strong northwesterly winds result in heavy snowfall on Mt. Tateyama (3015 m above sea level), which faces the Japan Sea. Aerosols are transported in a predictable manner by westerly wind from continental Asia to Japan and are preserved in the snowfall without undergoing local contamination (Osada et al., 2004; Watanabe et al., 2011). Snow cover is therefore regarded as a product of continuous precipitation during the Asian dust season, and can be a useful source of aerosol particulates.

Here, we report a shipboard experiment undertaken in the Pacific Ocean to assess the impact of aerosol particulates, at levels typically experienced by oceanic surface waters on marine microbial communities. We used a combination of physicochemical analyses, direct cell counting and high-throughput sequencing approaches to identify changes in phytoplankton and heterotrophic microbial communities.

2. Materials and methods

2.1. Aerosol particulate recovery

To prepare aerosol samples for shipboard experiments, snow samples containing aerosol particulates (dust particles and PM_{2.5} pollution particles) were collected from snow cover at Murododaira on Mt. Tateyama (36.57°N, 137.60°E; 2450 m, MR; Fig. 1) on 21

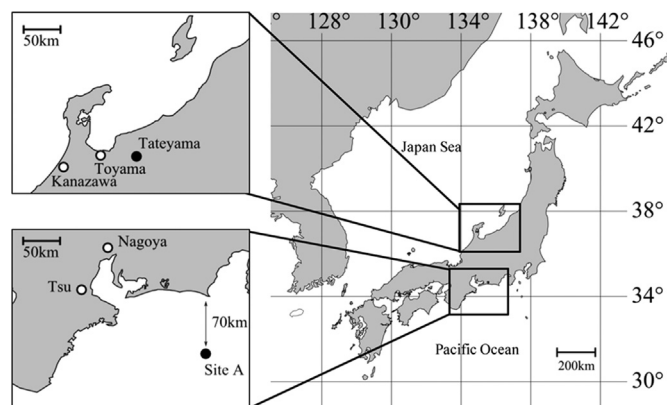


Fig. 1. Snow samples were collected from Murododaira (36.58°N, 137.60°E) on Mt. Tateyama. The shipboard incubation experiment was performed in the Pacific Ocean at Site A (34.07°N, 138.77°E).

March 2012. At this location, strong northwesterly winds result in heavy snowfall during the winter and early spring leading to the deposition of aerosol particulates originating from the Asian continent (Watanabe et al., 2011). After a snow pit was dug, the walls of the pit were carefully smoothed to ensure the stratigraphy of the snow layer was undisturbed. The snow layers were composed of compacted or solid-type snow and included layers of melted and refrozen water (ice). In particular, the layer from the snow surface to a depth of 150 cm were included in the brown-color layers (dirty layers) that had not melted. Therefore, these snow layers would be expected to essentially maintain the record of atmospheric aerosol deposition, revealing chemical compound dynamics from the top of the snow wall to a depth of 615 cm (Fig. S1). The snow layer at a depth of 110 cm was a non-white color (to be referred to as the “dirty” layer) and had higher concentrations of mineral particles (1.90×10^5 particles mL^{-1}) and calcium ($18.8 \mu\text{eq L}^{-1}$) than other snow layers. The dirty layer harbored Na^+ at a concentration of $13.8 \mu\text{eq L}^{-1}$, which primarily originated from sea salt. The contribution of sea salt Ca^{2+} to the total Ca^{2+} content was minor (approximately 3%). The solutions of snow samples from a depth of 110 cm contained nss- Ca^{2+} at a high concentration of $18.2 \mu\text{eq L}^{-1}$, suggesting that the mineral particles in the snow layers specifically contained calcium, which is a trace mineral found in Asian-dust mineral particles (Suzuki and Tsunogai, 1993). Snow was recovered aseptically from a subsurface of a dirty snow layer at a depth of 110 cm. We extrapolated LIDAR measurements of dust events and used back-trajectory analysis to establish the origin of this dust, deposited from the 1st to the 3rd of April 2012, in mainland Asia (Fig. S2). The snow samples were stored in polycarbonate bottles that were washed in a 3 mol L^{-1} HCl solution and sterilized at $121 \text{ }^\circ\text{C}$ for 20 min to remove metal, nitrate, and phosphate contamination, and frozen at $-80 \text{ }^\circ\text{C}$ until further analysis.

2.2. Seawater incubations

Seawater sampling and shipboard experiments were performed 70 km off the coast of Omaezaki in the Shizuoka prefecture (34.07°N, 138.77°E) on 26 May 2012 during the SE526 cruise of T/S Seisui Maru (Mie University) (Fig. 1). At this sampling site, the water column is thermally stratified from spring through summer, and the seawater temperature during the sampling periods decreased from $19.2 \text{ }^\circ\text{C}$ to $18.7 \text{ }^\circ\text{C}$ between a depth of 10 m and 18 m, indicating the presence of a weak thermocline (Fig. S3). These results suggest that the sampling area was oligotrophic on 26 May 2012. The nutrients in the surface layers are consumed by microbial activity, often causing oligotrophic conditions in the surface of the Pacific Ocean around Japan (Hashimoto et al., 2005). Seawater samples were collected from the surface at a depth of less than 1 m and stored in 20 L and 4 L sterilized holding tanks. One 20 L and two 4 L experimental tanks were used for each treatment, including ones filled with water as the control (C), ones with water exposed to 0.05 mg L^{-1} of aerosols simulating a light dust event (LS), and with water exposed to 0.25 mg L^{-1} of aerosols simulating a heavy dust event (HS). Treatments were incubated onboard the ship at surface temperature ($24 \text{ }^\circ\text{C}$) under ambient light conditions for a 72 h period. Initial measurements were conducted 30 min after addition of aerosol particulates, with 24 h sample intervals thereafter.

2.3. Water chemistry

For seawater samples, nitrate concentration was determined colorimetrically (Strickland and Parsons, 1972) after first removing particulates by filtration through $0.2 \mu\text{m}$ pore-size polycarbonate filters (Millipore, Tokyo, Japan). Inorganic phosphorus, as well as particulate and dissolved organic phosphorus, were measured using the molybdenum blue method (Strickland and Parsons, 1972). Iron in seawater samples was measured using ICP-AES.

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